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54 **Moldable reinforced composition containing styrenic resin.**

- 57 A styrene-based resin composition comprises by weight a kneaded mixture of
- (a) 100 parts of a blend of a styrene-based resin and a rubber-modified styrene-based resin, or a rubber-modified styrene-based resin;
 - (b) 1 to 45 parts of a bromine compound having a bromine content of 40 wt% or more and a melting point of 80°C or higher, e.g. a tetrabromobisphenol A carbonate oligomer;
 - (c) 0.5 to 15 parts of antimony trioxide powder; and
 - (d) 1 to 30 parts of glass fibers, e.g. of length 3-15 µm and diameter 0.5-6 mm;
- the heat distortion temperature of a molded product obtainable from said composition being at least 85° C and the impact strength thereof is 5.0 kg.cm/cm or more.
 A light discoloration preventing mixture (e) can be included.
- The composition is easily made and has excellent flame retardance, heat resistance, and impact resistance without impairing the inherent mechanical characteristics of the styrene based resin.
- The molded composition is cured, e.g. at 200°C; the molded products obtained are useful as armoring materials, of light electrical appliances or office machines.

EP 0 272 114 A1

MOLDABLE REINFORCED COMPOSITION CONTAINING STYRENIC RESIN

This invention relates to a reinforced styrene-based resin composition, more particularly to a styrene-based composition which has excellent flame retardance, heat resistance and impact strength without reduction in the inherent mechanical characteristics of a styrenic resin. The composition provides a material useful as armoring materials or the like, for e.g. light electrical appliances and office machines.

5 Styrene-based synthetic resins modified (reinforced with a rubbery elastomer form a material which is excellent in processability, mechanical properties, especially impact strength, and electrical properties and are in wide use as various molded products used in electrical industry.

However, such rubber-modified styrene-based resins are limited in their use because of their ready combustibility and comparatively low resistance to heat generated by or radiated from electrical circuits.

10 Therefore, in order to obviate this ready combustibility, there are known methods which impart flame retardance to styrene-based resins by incorporating therein various organic halogen compounds, phosphoric ester compounds or antimony compounds.

Although the addition of these flame-retarding agents to styrene-based resins made them flame-retardant, it also gave rise to such defects as impairment of thermal properties and mechanical properties.

15 In order to impart especially sufficient flame retardance, it is required to add a large amount of flame retarding agent, but in such a case, it is a defect that the heat resistance, impact resistance, etc. markedly decrease as the amount of the flame retarding agent added increases.

Further, the styrene-based resins having these flame-retarding agents added are very poor in lightfastness, so that in the case where they are used as an armoring material of light electrical appliances or office machines, they are liable to discolor by natural light or rays from artificial illumination, etc. and in particular with material which is toned to a white color or light color, and so they could be hardly as such.

This invention is intended to provide a flame-retardant styrene-based resin composition which is excellent in flame retardance and impact resistance, while alleviating the above described problems in the prior art.

25 According to the invention a flame-retardant styrene-based resin composition comprises

(a) 100 parts by weight of a blend of a styrene-based resin and a rubber-modified styrene-based resin, or a rubber-modified styrene-based resin;

(b) from 1 to 45 parts by weight of a bromine compound whose bromine content is 40 wt% or more and whose melting point is 80°C or higher;

30 (c) from 0.5 to 15 parts by weight of antimony trioxide; and

(d) from 1 to 30 parts by weight of glass fibers, wherein the heat distortion temperature of a molded product obtainable from said composition is 85° C or higher and the impact strength thereof is 5.0 kg.cm/cm or more.

The styrene-based resins used in the component (a) in this invention include not only polymers that can be obtained by homopolymerization of styrenic monomers (such as styrene, methylstyrene or chlorostyrene) such as polystyrene, polymethylstyrene or polychlorostyrene, but also copolymers of these styrenic monomers.

40 These styrene-based resins can be prepared by various polymerization processes such as bulk polymerization, solution polymerization, emulsion polymerization, suspension polymerization or bulk-suspension polymerization.

The rubber-modified styrene-based resins which are also used in the component (a) in this invention include copolymers of styrenic monomers (such as styrene, methylstyrene, chlorostyrene) and other copolymerizable monomers or elastomers.

45 As other monomers copolymerizable with the styrenic monomers, there can be mentioned, for example, acrylonitriles (such as acrylonitrile or methacrylonitrile); unsaturated carboxylic acids (such as acrylic acid or methacrylic acid); unsaturated carboxylic acid esters (such as methyl acrylate or methyl methacrylate); dienes (such as butadiene, isoprene or chloroprene); olefins (such as 1-butene or 1-pentene); and α,β - unsaturated carboxylic acids or anhydrides thereof (such as maleic anhydride).

50 Also, as the elastomers copolymerizable with the styrenic monomers, there can be mentioned polydiene rubbers (such as butadiene rubber, isoprene rubber or chloroprene rubber); styrene-diene elastomers (such as styrene-butadiene rubber); and block copolymer elastomers having a polybutadiene block in the molecular elastomers having a polybutadiene block in the molecular structure (such as styrene-butadiene-styrene rubber).

These rubber-modified styrene-based resins can also be prepared by the same various polymerization processes as for the above described styrene-based resins.

It is preferable that the blend of a styrene-based resin and a rubber-modified styrene-based resin or the rubber-modified styrene-based resin which is used as the component (a) has a heat distortion temperature of 75 °C or higher. If the heat distortion temperature is lower than 75° C, it is difficult to obtain flame-retardant styrene-based resin composition having the required heat distortion temperature of 85°C or higher.

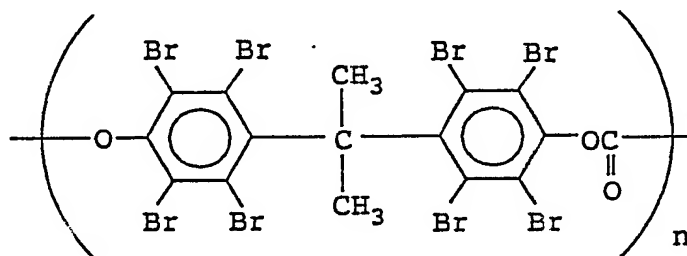
Further, as the base resin component (a), a blend of a styrene-based resin and a rubber-modified styrene-based resin or a rubber-modified styrene-based resin may be used alone, or each can be a mixture of two or more thereof selected independently and in any desired proportion.

The heat distortion temperature, which is used as an index of heat resistance in this specification, is measured at a fiber stress of 18.6 kg/cm² according to JIS K7207.

The impact strength is measured on a notched specimen of which a molded product has a thickness of 2 mm according to ASTM D256.

The bromine compounds used as a flame-retarding agent used as the component (b) in this invention, the bromine content is 40 wt% or more, preferably 50 wt% or more, and more preferably 60 wt% or more; and the melting point is 80°C or higher, preferably 150°C or higher, and more preferably 250°C or higher.

As the bromine compounds, there can be mentioned, for example, decabromodiphenyl oxide, octabromodiphenyl oxide, tetrabromobisphenol A, tetrabromobisphenol A-bis-(2,3-dibromopropyl) oxide, tetrabromobisphenol S, bis(tribromophenoxy)ethane. A tetrabromobisphenol A carbonate oligomer represented by the following formula is also a preferable bromine compound:



wherein n represents an integer of from 3 to 20.

In the above general formula, when n is larger than 20, though the heat resistance of the flame-retardant styrene-based resin composition can be improved, the impact strength is lowered; and when n is smaller than 3, though the impact strength is good, the heat resistance is not improved, so that these cases are all undesirable.

The bromine compounds are not limited to those mentioned above, and they may be used singly or in combination of two or more thereof which are taken simultaneously.

In the case where the bromine content in the bromine compound is less than 40 wt%, in order to impart the flame retardance, a larger amount of the bromine compound is required to be incorporated, so that the impact resistance of the resin composition is undesirably degraded.

Also, in the case where the melting point of the bromine compound is lower than 80°C, the heat resistance of the resin composition is much reduced, so that this is not desirable.

The amount of the bromine compound (b) to be incorporated as the flame retarding agent in this invention is from 1 to 45 parts by weight and preferably from 2 to 30 parts by weight on the basis of 100 parts by weight of the blend of a styrene-based resin and a rubbermodified styrene-based resin, or the rubber-modified styrene-based resin as the base resin (a).

When the amount of the flame retarding agent incorporated is less than 1 part by weight, the flame retardance of the styrene-based resin composition is scarcely improved at all, while when it exceeds 45 parts by weight, the mechanical strengths, such as impact strength, are undesirable degraded.

Antimony trioxide used as the component (c) in this invention is a usually commercially available fine powder thereof and can be used in combination with a flame retarding agent in a mixing ratio of the flame retarding agent to the antimony trioxide being from 1/10 to 10/1 by weight.

The amount of antimony trioxide to be incorporated in this invention is from 0.5 to 15 parts by weight on the basis of 100 parts by weight of the blend of a styrene-based resin and a rubber-modified styrene-based resin, or the rubber-modified styrene-based resin. In the case where the amount of antimony trioxide incorporated is less than 0.5 part by weight, the flame retardance of the styrene-based resin composition obtained almost cannot be improved, while in the case where it exceeds 15 parts by weight, the mechanical strengths, such as, for example, impact strength, are undesirably degraded.

Next, the glass fibers used as the component (d) in this invention are those of which the fiber diameter is 20 μm or less and preferably from 3 to 15 μm , and the fiber length is from 0.3 to 10 mm and preferably from 0.5 to 6 mm.

When the fiber diameter of the glass fibers exceeds 20 μm , the external appearance of the molded product of the composition undesirably becomes uneven, and also, when the fiber length of the glass fibers is less than 0.3 mm, the improvements in the heat resistance and the impact resistance of the resin composition are insufficient, and when exceeding 10 mm, the external appearance of the molded product undesirably becomes uneven.

The glass fibers are preferably those which have improved adhesion against the styrene-based resin and/or rubber-modified styrene-based resin by treating the surface thereof with an aminosilane-based coupling agent and then binding with an acrylic resin.

The amount of the glass fibers to be incorporated in this invention is from 1 to 30 parts by weight and preferably from 5 to 25 parts by weight on the basis of 100 parts by weight of the blend of a styrene-based resin and a rubber-modified styrene-based resin, or the rubber-modified styrene-based resin. The larger the amount of the glass fibers incorporated, the higher the heat resistance and the impact resistance, but when exceeding 30 parts by weight, the impact resistance may sometimes decrease on the contrary, and the external appearance of the molded product may also become poor, so that these are not desirable. On the other hand, when the amount of the glass fibers incorporated is less than one part by weight, the heat resistance of the resin composition cannot be sufficiently improved.

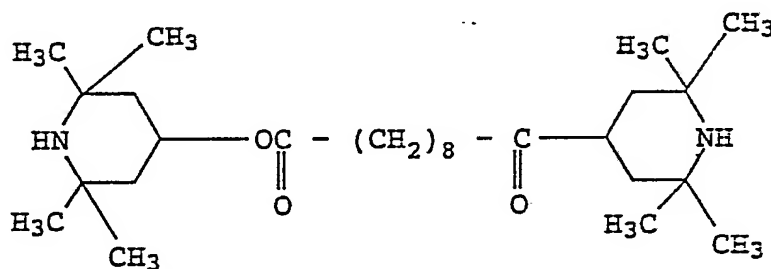
Further, the compositions may include (e) light discoloration inhibitors when required. As such light discoloration inhibitors, ultraviolet light absorbents, ultraviolet light stabilizers, and coloration preventing agents can be used.

The ultraviolet light absorbent used as the light discoloration inhibitor (e) in this invention can be any one chosen arbitrarily from benzotriazole compounds, benzophenone compounds, benzoate compounds and cyanoacrylate compounds, and preferably benzotriazole compounds such as, for example, 2-(2'-hydroxy-3'-t-butyl-5'-methylphenyl)-5-chlorobenzotriazole (UVA-1).

The amount of the ultraviolet light absorbent to be incorporated is from 0.005 to 1.0 part by weight and preferably from 0.01 to 0.5 part by weight on the basis of 100 parts by weight of the blend of a styrene-based resin and a rubber-modified styrene-based resin, or the rubber-modified styrene-based resin.

When the amount of the ultraviolet light absorbent is less than 0.005 part by weight, the light discoloration resistance of the resin composition cannot be sufficiently improved, whereas when exceeding 1.0 part by weight, the inherent mechanical properties of the resin composition undesirably lower.

The ultraviolet light stabilizer used as the light discoloration inhibitor (e) in this invention includes hindered amine compounds such as, for example, a compound represented by the following structural formula:



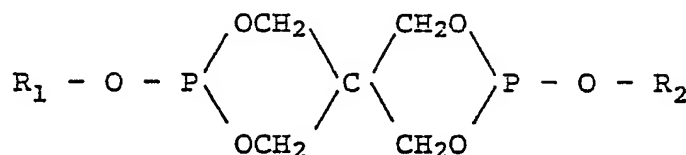
(UVS-1)

The amount of the hindered amine compound incorporated as the ultraviolet light stabilizer is from 0.005 to 1.0 part by weight and preferably from 0.01 to 0.5 part by weight on the basis of 100 parts by weight of the blend of a styrene-based resin and a rubber-modified resin, or the rubber-modified styrene-based resin.

When the amount of the hindered amine compound incorporated is less than 0.005 part by weight, the light discoloration resistance of the resin composition cannot be sufficiently improved, whereas when exceeding 1.0 part by weight, the inherent mechanical properties of the resin composition are impaired.

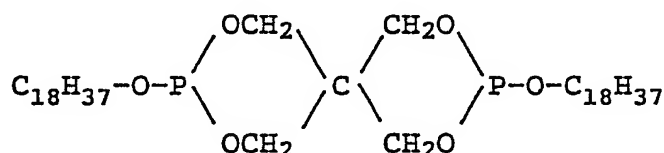
The coloration preventing agent used as the light discoloration inhibitor (e) in this invention includes

polymeric organophosphate compounds such as, for example, a compound represented by the following formula:

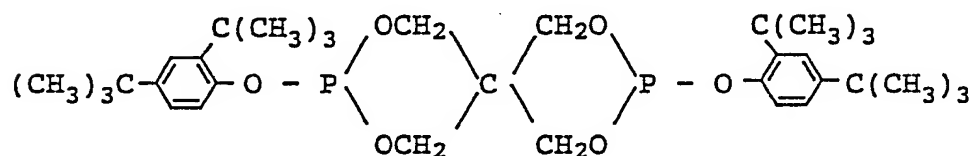


wherein R_1 and R_2 each represents a hydrogen atom, an alkyl group, an aryl group, a cycloalkyl group, an arylalkyl group or an alkylaryl group.

Specific examples of compounds represented by the above described general formula may be shown by the following rational formulae:



(CL-1)



(CL-2)

The amount of the coloration preventing agent to be incorporated in this invention is from 0.005 to 1.0 part by weight and preferably from 0.01 to 0.5 part by weight on the basis of 100 parts by weight of the blend of a styrene-based resin and a rubber-modified styrene-based resin, or the rubber-modified styrene-based resin, and can be used in combination with the above described ultraviolet light absorbers and ultraviolet light stabilizers.

When the amount of the coloration preventing agent incorporated is less than 0.005 part by weight, the light discoloration resistance of the resin composition cannot be sufficiently improved, whereas when exceeding 1.0 part by weight, the inherent mechanical properties of the resin composition lower.

In this invention, besides the above described light discoloration inhibitors, those which can serve as acceptors of the halogen set free from flame-retarding agents at the time of irradiation of ultraviolet light or at high temperatures, e.g., basic materials such as, for example, magnesium hydroxide, hydrotalcites, etc., can also be incorporated so as to still further improve the light discoloration resistance.

In the flame-retardant styrene-based resin composition of this invention, in addition to the above described components, commonly used coloring agents or lubricants such as stearic acid or stearates, can further be incorporated as occasion demands.

Now, with reference to some examples, this invention will be explained more fully below, and in these examples the evaluation of the resins as well as the resin compositions was made according to the following scheme.

(1) Melt flow index (MFI): Measured at 200°C and 5 kg according to JIS K7210.

(2) The heat distortion temperature as the measure of evaluating heat resistance was measured at a fiber stress of 18.6 kg/cm² according to JIS K7207.

(3) The Izod impact strength as the measure of evaluating impact strength was measured with respect to a notched specimen having a thickness of 2 mm according to ASTM D256.

(4) Flame retardance: The vertical or horizontal combustion test prescribed in U.S. UL Standard, UL 94 was carried out with respect to a test piece having a thickness of 2.0 mm.

(5) As the measure of evaluating light discoloration inhibition, the following measurement was made. After a molded product to be tested had been irradiated for 40 hours at a black panel temperature of 63°C without water spraying using a sunshine superlong life weatherometer (WEL-SUN-DCH type; manufactured



by Suga Testing Machine Co., Ltd.), the color difference (ΔE) between the irradiated specimen and the non-irradiated was measured by means of an SM Color Computer (SM-3 type: manufactured by Suga Testing Machine Co., Ltd.).

The smaller the color difference (ΔE), the better, but the criterion of the light discoloration inhibition was made as follows.

O: $\Delta E \leq 3$

x: $\Delta E > 3$

10 EXAMPLES 1 to 5 and COMPARATIVE EXAMPLES 1 to 5

Resin compositions were prepared by mixing each of the following components in the ratio shown in Table 1 and then kneading the resulting mixture at 190 to 220°C.

Molded products were obtained by injection molding the compositions at a molding temperature of 200°C.

The results of the measurement of the physical properties of the thus obtained molded products are shown in Table 1.

20 Styrene-based resin:

Esbrite®; manufactured by Nippon Polystyrene Industry Co., Ltd.

As general grade (GP) #8: heat distortion temperature = 90°C, Izod impact strength = 1.7 kg•cm/cm, MFI = 3 g/10 min.

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Rubber-modified styrene-based resin:

Esbrite®; manufactured by Nippon Polystyrene Industry Co., Ltd.

30 As impact resistant grade (HI 500 HM): heat distortion temperature = 84°C, Izod impact strength = 7.0 kg•cm/cm, MFI = 3 g/10 min.

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Flame retarding agents used:

10 DBDPO: Decabromodiphenyl oxide; manufactured by Dead Sea
Bromine Co., Ltd. in Israel.

15 Trade Name: FR1210

Bromine content: 82%

Melting point: 300°C

20 TBA: Tetrabromobisphenol A; manufactured by Teijin
Chemicals Ltd.

25 Trade Name: Fire Guard® 2000

Bromine content: 58%

Melting point: 180°C

30 TBA-DBPO: Tetrabromobisphenol A-dibromopropyl oxide;
manufactured by Teijin Chemicals Ltd.

35 Trade Name: Fire Guard® 3100

Bromine content: 67.8%

Melting point: 90-105°C

40 TBS: Tetrabromobisphenol S; manufactured by Matsunaga
Chemical Industries Co., Ltd.

45 Bromine content: 55%

Melting point: 290°C.

Antimony trioxide used: TWINKLING STAR Mark; produced in
50 Hunan, People's Republic of China

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Product of high purity, containing 99.6% or more
of antimony trioxide.

Glass fibers used:

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A: ECS-015-T321; manufactured by Nippon Electric
Glass Co., Ltd.

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Fiber diameter 13 microns, fiber length 1.5 mm,
processed with aminosilane-treated acrylic binder.

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B: ECS-03-T321; manufactured by Nippon Electric Glass
Co., Ltd.

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Fiber diameter 13 microns, fiber length 3 mm,
processed with aminosilane-treated acrylic binder.

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TABLE 1

Resin Composition (mixture ratio)														
Base resin			Rubber-modified					Results of evaluation						
	Styrene-based resin	Kind	Part by weight	styrene-based resin		Flame retarding agent		Antimony trioxide	Part by weight	Glass fibers	Part by weight	Flame retardance	Heat Distortion temperature (°C)	Izod impact strength (kg·cm/cm)
				Kind	Part by weight	Kind	Part by weight							
Example 1	GP #8	50	500 HM	50	DBDPO	13	4	A	8	V-0	90	6.0		
"	"	50	"	50	"	13	4	A	5	V-0	90	5.5		
"	"	50	"	50	TBA-OBPO	10	3	A	20	V-2	87	6.9		
"	-	-	"	100	TBS	15	5	A	8	V-2	88	5.0		
"	-	-	"	100	TBA	25	5	B	30	V-0	87	5.2		
Comparative Example 1	GP #8	50	500 HM	50	DBDPO	13	0.3	A	20	HB	90	6.5		
"	"	50	"	50	"	13	4	A	0.5	V-0	83	5.8		
"	"	100	"	0	"	13	4	A	25	V-0	94	3.9		
"	"	50	"	50	"	47	10	A	10	V-0	83	4.0		
"	"	50	"	50	"	13	4	A	35	V-2	97	4.6		

EXAMPLES 6 TO 8 AND COMPARATIVE EXAMPLES 6 AND 7

Resin pellets were prepared by mixing each of the following components in the ratio as shown in Table 2 and then kneading the resulting mixture at 190 to 220°C.

Then molded products were obtained by injection molding and their physical properties were measured, with the results being shown in Table 2.

Styrene-based resin

Esbrite®; manufactured by Nippon Polystyrene Industry Co., Ltd.

As general grade (GP) #8: heat distortion temperature = 90°C, Izod impact strength = 1.7 kg•cm/cm, MFI = 3 g/10 min.

Rubber-modified styrene-based resin:

Esbrite®; manufactured by Nippon Polystyrene Industry Co., Ltd.

As impact resistant grade (HI 500 HM): heat distortion temperature = 84°C, Izod impact strength = 7.0 kg•cm/cm, MFI = 3 g/10 min.

As superimpact resistant grade (HI 500 SB): heat distortion temperature = 82°C, Izod impact strength = 12 kg•cm/cm, MFI = 2.2 g/10 min.

Flame retarding agent used:

Tetrabromobisphenol A carbonate oligomer; manufactured by Teijin Chemicals Ltd.

Trade Name: Fire Guard® 7500

Bromine content: 50.6%

Melting point: 220°C.

Antimony trioxide used:

TWINKLING STAR Mark; produced in Hunan, People's Republic of China.

Product of high purity, containing 99.6% or more of antimony trioxide.

Glass fibers used:

ECS-015-T321; manufactured by Nippon Electric Glass Co., Ltd.

Fiber diameter 13 microns, fiber length 1.5 mm, processed with aminosilane-treated acrylic binder.

Light discoloration inhibitor used:

The case denoted by light discoloration inhibitor "present" shows that 0.2 part by weight of ultraviolet light absorbent (a benzotriazole compound; Sumisorb® 300 (UVA-1); manufactured by Sumitomo Chemical Company, Limited), 0.1 part by weight of ultraviolet light stabilizer (a hindered amine compound; Sanol® LS770 (UVS-1) manufactured by Ciba-Geigy (Japan) Limited), and 0.05 part by weight of coloration preventing agent (a polymeric organophosphate compound; Ultrinox® 626 (CL-2) manufactured by Borg-Warner Corp.) were incorporated. The case denoted by light discoloration inhibitor "absent" shows that the above described mixture of ultraviolet light absorbent, ultraviolet light stabilizer and coloration preventing agent was not incorporated.

TABLE 2

Resin composition (mixing ratio)														
Base resin			Rubber-modified styrene-based resin		Flame retarding agent		Antimony trioxide		Glass fiber		Light discoloration inhibitor			
Styrene-based resin	part by Kind	weight	part by Kind	weight	part by weight	part by weight	part by weight	part by weight	part by weight	part by weight	Flame resistance	Results of evaluation		
											Light coloration resistance	Heat distortion temperature (°C)	Izod impact strength (kg·cm/cm)	
Example 6	-	-	500 HM	100	18	4.5	8	present			V-0	○	94	6.0
"	7 GP#8	20	"	80	18	4.5	10	present			V-0	○	94	5.2
"	8	-	500 HM	60	21	5	8	present			V-0	○	89	7.2
			500 SB	40										
Compara-	-	-	500 HM	100	18	4.5	8	absent			V-0	×	94	6.1
tive														
Example 6														
"	7	-	500 HM	60	21	5	8	absent			V-0	×	90	7.2
			500 SB	40										

As is readily understood from the foregoing description, the flame-retardant styrene-based resin composition of this invention is excellent in flame retardance, heat resistance and impact resistance without impairing inherent mechanical characteristics of the styrene-based resin.

Further, each of the components which are incorporated in the flame-retardant styrene-based resin composition of this invention are not only easy to handle but also nonpoisonous, so that the molded products obtained from the composition are useful as armoring materials or the like, of light electrical appliances, office machines and similar products.

Claims

1. A flame-retardant styrene-based resin composition which comprises:

(a) 100 parts by weight of a blend of a styrene-based resin and a rubber-modified styrene-based resin, or a rubber-modified styrene-based resin;

(b) from 1 to 45 parts by weight of a bromine compound having a bromine content of 40 wt% or more and a melting point of 80°C or higher;

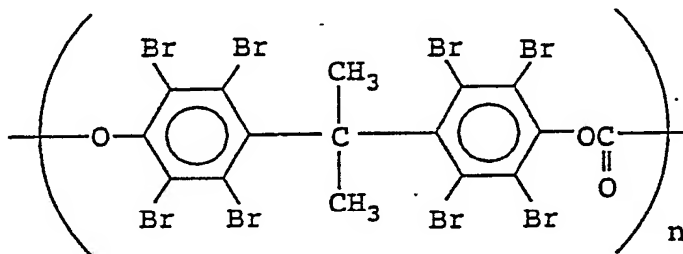
(c) from 0.5 to 15 parts by weight of antimony trioxide; and

(d) from 1 to 30 parts by weight of glass fibers, wherein the heat distortion temperature of a molded product obtainable from said composition is 85°C or higher and the impact strength thereof is 5.0 kg•cm/cm or more.

2. A flame-retardant styrene-based resin composition as in Claim 1, wherein the heat distortion temperature of the component (a) is 75°C or higher.

3. A flame-retardant styrene-based resin composition as in Claim 1 or 2, wherein the component (b) is at least one of decabromodiphenyl oxide, octabromodiphenyl oxide, tetrabromobisphenol A, tetrabromobisphenol A-bis(2,3-dibromopropyl) oxide, bis(tribromophenoxy)ethane and tetrabromobisphenol S.

4. A flame-retardant styrene-based resin composition as in Claim 1 or 2, wherein bromine compound (b) is a tetrabromobisphenol A carbonate oligomer represented by the following formula:



wherein n represents an integer of 3 to 20.

5. A flame-retardant styrene-based resin composition as in any of Claims 1 to 4, wherein said resin composition also contains as a light discoloration inhibitor.

(i) from 0.005 to 1.0 part by weight of an ultraviolet light absorbent,

(ii) from 0.005 to 1.0 part by weight of an ultraviolet light stabilizer, and

(iii) from 0.005 to 1.0 part by weight of a coloration preventing agent.



EP 87 31 1103

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl. 4)
X	US-D- 519 095 (E.J. McINERNEY et al.) * Claims 1-7 * ---	1,3	C 08 L 25/04 C 08 K 13/04 //
X	DE-A-2 642 094 (CHEMISCHE FABRIK KALK GmbH) * Claims 1,3,6; example 6 * ---	1,3	(C 08 K 13/04 C 08 K 3:22 C 08 K 5:06 C 08 K 5:10 C 08 K 5:13 C 08 K 7:14)
X	PATENT ABSTRACTS OF JAPAN, vol. 6, no. 135 (C-115)[1013], 22nd July 1982; & JP-A-57 61 051 (UNITIKA K.K.) 13-04-1982 ---	1,3	
A	FR-A-2 292 006 (GENERAL ELECTRIC CO.) * Claim 1 * -----	1	
			TECHNICAL FIELDS SEARCHED (Int. Cl.4)
			C 08 K 13/00 C 08 K 3/00 C 08 K 5/00 C 08 K 7/00
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 23-02-1988	Examiner GLANDDIER A.
CATEGORY OF CITED DOCUMENTS X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document			